

Problems and paradoxes of the Lifshitz theory

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Abstract. The problems and paradoxes of the Lifshitz theory in application to real dielectric and semiconductor materials are reviewed. It is shown that the inclusion of drift current of conduction electrons into the model of dielectric response results in contradictions with both thermodynamics and experimental data of different experimental groups. Physical reasons why the problems and paradoxes arise are analyzed and found to be connected with the violation of basic applicability condition of the Lifshitz theory, the thermal equilibrium. A recent alternative approach to the resolution of the problems based on the inclusion of screening effects and diffusion current is considered and demonstrated to be thermodynamically and experimentally inconsistent. It is argued that the inclusion of the diffusion current leads to an even deeper violation of thermal equilibrium. Phenomenologically, the Lifshitz theory with role of drift and diffusion currents neglected is shown to be free of problems and in agreement with both thermodynamics and all available experimental data.

1. Introduction

It is common knowledge that the Lifshitz theory gives a description of the van der Waals and Casimir forces between two plane parallel plates (semispaces) in terms of the frequency-dependent dielectric permittivities of the plate materials $\varepsilon(\omega)$ [1–3]. Starting in 2000, problems and paradoxes with the application of the Lifshitz theory to real materials (at first to metals [4] and then to dielectrics [5]) were recognized. Specifically, for metals described by the Drude dielectric function a large thermal effect was predicted at short separations [4] in qualitative disagreement with the case of ideal metals. This prediction was excluded experimentally at a 99.9% confidence level [6,7]. At large separations, for the Drude metals one half of the Casimir force acting between ideal metals was obtained [4]. The same result was obtained in [8,9] by modelling a metal with a set of nonrelativistic hard-core or point-like particles confined by walls. In [10] it was shown that the substitution of the Drude dielectric function into the Lifshitz formulas results in a violation of the third law of thermodynamics for metals with perfect crystal lattices. In [11] it was demonstrated that for metals with impurities the Nernst heat theorem is satisfied. This, however, does not solve the problem because in accordance with quantum statistical physics the Nernst theorem must be satisfied for metals with perfect crystal lattices as well. The situation is even more dramatic for dielectrics, the materials for which the Lifshitz theory was originally formulated. It was shown that the inclusion of the dc conductivity arising in all dielectrics at nonzero temperature radically changes the theoretical predictions and leads to disagreement between the Lifshitz theory and thermodynamics [5,12,13].

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In this paper we review recent paradoxical applications of the Lifshitz theory to real materials (mostly to dielectrics) and analyze the physical reasons of why the paradoxes appear. We show that difficulties arise each time when the drift or diffusion currents are included in the model of the dielectric response. In this connection we discuss recent experiments with semiconductors [14,15] and dielectric [16] test bodies and show that the measurement results are in disagreement with theory taking the dc conductivity into account. Recent theoretical approaches attempting to solve problems of the Lifshitz theory by accounting for screening effects and diffusion currents [17,18] are demonstrated to be in contradiction with both thermodynamic and experimental data.

Our analysis leads to the conclusion that the roots of the problems are in the violation of the applicability conditions of the Lifshitz theory when the role of free charge carriers is included in the model of the dielectric response. We argue that in this case one violates the condition of thermal equilibrium by admitting a unidirectional flux of heat from the Casimir plates to the heat reservoir.

In Sec. 2 we discuss the consistency of the Lifshitz theory with thermodynamics. Sec. 3 is devoted to the experimental tests of the Lifshitz theory under different assumptions about the model of the dielectric response. Sec. 4 contains the formulation of a phenomenological approach of how to apply theory in agreement with both thermodynamics and the experimental data. In Sec. 5 we provide thermodynamic and experimental tests for the proposed modification of the reflection coefficients due to screening effects and diffusion current. Our discussion and conclusions are presented in Sec. 6.

2. Consistency of the Lifshitz theory with thermodynamics

The main result of the Lifshitz theory is the expression for the free energy of dispersion interaction per unit area of two semispaces at a separation distance a , at temperature T in thermal equilibrium with an environment [1–3]

$$\mathcal{F}(a, T) = \frac{k_B T}{2\pi} \sum_{l=0}^{\infty}{}' \int_0^{\infty} k_{\perp} dk_{\perp} \left\{ \ln \left[1 - r_{\text{TM}}^{(1)}(i\xi_l, k_{\perp}) r_{\text{TM}}^{(2)}(i\xi_l, k_{\perp}) e^{-2aq_l} \right] \right. \\ \left. + \ln \left[1 - r_{\text{TE}}^{(1)}(i\xi_l, k_{\perp}) r_{\text{TE}}^{(2)}(i\xi_l, k_{\perp}) e^{-2aq_l} \right] \right\}. \quad (1)$$

Here, k_{\perp} is the magnitude of a wave vector in the plane of boundary planes (perpendicular to the z -axis), prime adds a multiple $1/2$ to the term $l = 0$, $\xi_l = 2\pi k_B T l / \hbar$ ($l = 0, 1, 2, \dots$) are the Matsubara frequencies, k_B is the Boltzmann constant, and the Fresnel reflection coefficients along the imaginary frequency axis are given by ($n = 1, 2$)

$$r_{\text{TM}}^{(n)}(i\xi_l, k_{\perp}) = \frac{\varepsilon_l^{(n)} q_l - k_l^{(n)}}{\varepsilon_l^{(n)} q_l + k_l^{(n)}}, \quad r_{\text{TE}}^{(n)}(i\xi_l, k_{\perp}) = \frac{q_l - k_l^{(n)}}{q_l + k_l^{(n)}}, \quad (2)$$

where

$$q_l^2 = k_{\perp}^2 + \frac{\xi_l^2}{c^2}, \quad k_l^{(n)2} = k_{\perp}^2 + \varepsilon_l^{(n)} \frac{\xi_l^2}{c^2}, \quad \varepsilon_l^{(n)} \equiv \varepsilon^{(n)}(i\xi_l). \quad (3)$$

The free energy of atom-wall interaction, where $\varepsilon_l^{(1)}$ is the dielectric permittivity of wall material, can be expressed as

$$\mathcal{F}^A(a, T) = -k_B T \sum_{l=0}^{\infty}{}' \alpha(i\xi_l) \int_0^{\infty} k_{\perp} dk_{\perp} q_l e^{-2aq_l} \\ \times \left\{ 2r_{\text{TM}}^{(1)}(i\xi_l, k_{\perp}) - \frac{\xi_l^2}{q_l^2 c^2} \left[r_{\text{TM}}^{(1)}(i\xi_l, k_{\perp}) + r_{\text{TE}}^{(1)}(i\xi_l, k_{\perp}) \right] \right\}, \quad (4)$$

where $\alpha(i\xi_l)$ is the dynamic atomic polarizability.

For dielectrics, the dielectric permittivity along the imaginary frequency axis is commonly presented in the form [19]

$$\varepsilon(i\xi) = 1 + \sum_j \frac{g_j}{\omega_j^2 + \xi^2 + \gamma_j \xi} \approx 1 + \sum_j \frac{g_j}{\omega_j^2 + \xi^2}, \quad (5)$$

where $\omega_j \neq 0$ are the oscillator frequencies, g_j are the oscillator strength and γ_j are the relaxation parameters. This representation assumes some finite value of the static dielectric permittivity

$$\varepsilon_0 \equiv \varepsilon(0) = 1 + \sum_j \frac{g_j}{\omega_j^2} < \infty. \quad (6)$$

Using this model one can find the asymptotic behavior of the free energy of the Casimir interaction between two similar dielectric plates with $\varepsilon^{(1)} = \varepsilon^{(2)} = \varepsilon$ at low temperature $T \ll T_{\text{eff}} \equiv \hbar c / (2ak_B)$ [5]

$$\mathcal{F}(a, T) \approx E(a) - \frac{\hbar c}{32a^3} \zeta(3) r_0^2 (\varepsilon_0 + 1) \left(\frac{T}{T_{\text{eff}}} \right)^3. \quad (7)$$

Here, $E(a)$ is the Casimir energy at zero temperature, $\zeta(z)$ is the Riemann zeta function, and $r_0 \equiv (\varepsilon_0 - 1)/(\varepsilon_0 + 1)$ is the value of the TM reflection coefficient at zero frequency. A similar asymptotic expression can be obtained from (4) for the free energy of the atom-wall interaction [20]

$$\mathcal{F}^A(a, T) \approx E^A(a) - \frac{\hbar c \pi^3}{240a^4} \alpha(0) C_D(\varepsilon_0) \left(\frac{T}{T_{\text{eff}}} \right)^4, \quad (8)$$

where $C_D(\varepsilon_0)$ is some constant found in [20].

Equations (7) and (8) allow one to find the asymptotic behavior of the Casimir entropy in the plate-plate and atom-plate configurations

$$S(a, T) = -\frac{\partial \mathcal{F}(a, T)}{\partial T}, \quad S^A(a, T) = -\frac{\partial \mathcal{F}^A(a, T)}{\partial T} \quad (9)$$

at low temperature. Thus, from (7) and (8) one obtains

$$\begin{aligned} S(a, T) &\approx \frac{3k_B}{16\pi a^2} \zeta(3) r_0^2 (\varepsilon_0 + 1) \left(\frac{T}{T_{\text{eff}}} \right)^2, \\ S^A(a, T) &\approx \frac{\pi^3 k_B}{30a^3} \alpha(0) C_D(\varepsilon_0) \left(\frac{T}{T_{\text{eff}}} \right)^3. \end{aligned} \quad (10)$$

As is seen from (10), in both cases the Casimir entropy goes to zero when T vanishes, i.e., the Lifshitz theory with the dielectric permittivity (5) satisfies the Nernst heat theorem.

The model of the dielectric response (5) is, however, not exact. It does not take into account that at $T \neq 0$ all dielectric materials possess small but physically real dc conductivity $\sigma(T)$. For all dielectric materials, the general behavior of $\sigma(T)$ with vanishing temperature is given by

$$\sigma(T) \sim \exp\left(-\frac{C}{k_B T}\right) \rightarrow 0 \quad \text{when } T \rightarrow 0, \quad (11)$$

where the physical meaning of the coefficient C is different for different classes of dielectrics [21,22]. The dc conductivity of dielectrics is usually taken into account by means of the Drude-like term (see, e.g., [23])

$$\tilde{\varepsilon}(i\xi, T) = \varepsilon(i\xi) + \frac{4\pi\sigma(T)}{\xi}. \quad (12)$$

It would be reasonable to expect that the substitution of the dielectric permittivity (12), instead of (5), into the Lifshitz formulas (1) and (4) leads to only minor corrections to the asymptotic results (7) and (8). In reality, however, this substitution radically changes the asymptotic behavior of the free energy at low temperature:

$$\begin{aligned}\tilde{\mathcal{F}}(a, T) &= \mathcal{F}(a, T) - \frac{k_B T}{16\pi a^2} \left[\zeta(3) - \text{Li}_3(r_0^2) + R(T) \right], \\ \tilde{\mathcal{F}}^A(a, T) &= \mathcal{F}^A(a, T) - \frac{k_B T}{4a^3} \left[(1 - r_0)\alpha(0) + \tilde{R}(T) \right].\end{aligned}\quad (13)$$

Here, $\text{Li}_n(z)$ is the polylogarithm function, $\mathcal{F}(a, T)$ and $\mathcal{F}^A(a, T)$ are given by (7) and (8), respectively, and $R(T)$, $\tilde{R}(T)$ are exponentially vanishing functions when $T \rightarrow 0$. In this case the definition of the Casimir entropy (9) results in the following nonzero values at zero temperature [5,20]

$$\begin{aligned}\tilde{S}(a, 0) &= \frac{k_B}{16\pi a^2} \left[\zeta(3) - \text{Li}_3(r_0^2) \right] > 0, \\ \tilde{S}^A(a, 0) &= \frac{k_B}{4a^3} (1 - r_0)\alpha(0) > 0.\end{aligned}\quad (14)$$

This is in violation of the third law of thermodynamics (the Nernst theorem) because not only are these values not equal to zero, but they also depend on the parameters of the system (separation distance, static dielectric permittivity and atomic polarizability).

The obtained result can be considered as a puzzle because instead of more accurate asymptotic expressions one arrives to the conclusion of an apparent contradiction of the Lifshitz theory with the fundamental principle of thermodynamics.

3. Experimental tests

Rapid progress in the measurements of the Casimir force is very helpful for the resolution of the theoretical controversies. As mentioned in the Introduction, the experiment [6,7] excluded the use of the Drude model to describe the thermal Casimir force between metal plates. It is important to note, however, that a much smaller thermal effect, as predicted by the generalized plasma model [24,25], was not registered in that experiment. Here we consider the results of two other experiments involving semiconductor and dielectric materials [14–16]. Both of them are accurate enough to resolve between the theoretical approaches including and neglecting the dielectric dc conductivity. What is more, one of them [16] provides the first experimental observation of the thermal effects in Casimir physics.

We start with the observation of modulation of the Casimir force between a gold coated sphere and a Si plate with laser light [14,15]. In this experiment, a B doped p -type Si plate was illuminated with laser pulses and the difference of the Casimir forces between a sphere and a plate in the presence and in the absence of light has been measured. In the absence of laser pulse the concentration of charge carriers in the Si plate was $\tilde{n} = 5 \times 10^{14} \text{ cm}^{-3}$ and in the presence of light $n_1 = 2.1 \times 10^{19} \text{ cm}^{-3}$ or $n_2 = 1.4 \times 10^{19} \text{ cm}^{-3}$ for the two different absorbed powers $P_{w1} = 9.3 \text{ mW}$ and $P_{w2} = 4.7 \text{ mW}$. The directly measured quantity was, thus, $\Delta F(a) = F^L(a) - F(a)$, where F^L and F are the Casimir forces in the presence and in the absence of laser light on the plate, respectively. The experimental data were compared with the Lifshitz theory either neglecting or including dc conductivity of the high-resistivity Si in the dark phase.

In Fig. 1(a,b) the mean experimental data $\langle \Delta F(a) \rangle$ are shown as dots for the absorbed powers P_{w1} and P_{w2} , respectively. The calculational results for $\Delta F(a)$, using the Lifshitz theory with neglect of the dc conductivity of the high-resistivity Si (dark phase), are shown as the solid

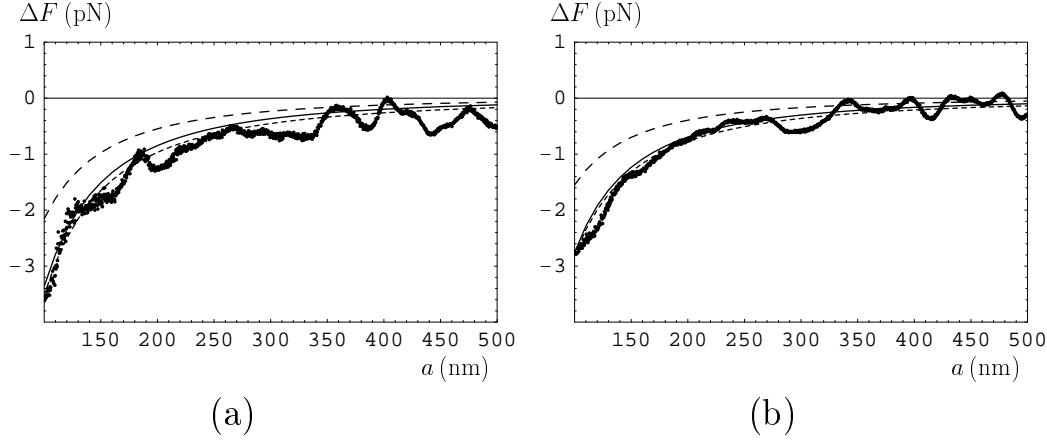


Figure 1. Differences of the Casimir forces in the presence and in the absence of light versus separation for different absorbed powers: (a) 9.3 mW and (b) 4.7 mW. The mean measured differences are shown as dots. Differences calculated using the Lifshitz formula at $T = 300$ K and $T = 0$ are shown as the solid and short-dashed lines, respectively, and those calculated including the dc conductivity of high-resistivity Si as the long-dashed lines.

lines. Similar calculational results using the Lifshitz theory with included dc conductivity (12) in the dark phase are shown as the long-dashed lines. As is seen in Fig. 1(a,b) the solid lines are consistent with the data whereas the long-dashed lines are excluded over the wide separation region. In the same figure, the short-dashed lines show the computational results obtained from the Lifshitz formula at zero temperature. As is seen in the figure, the computational results at $T = 0$ are consistent with the data. These results do not depend on whether the dc conductivity was included or neglected. The experiment under consideration is precise enough to exclude the large thermal effect, as obtained when the dc conductivity in the dark phase is included, but is not of sufficient precision to measure the small thermal effect obtained in the case when the dc conductivity in the dark phase is neglected.

For illustrative purpose, in Fig. 2 we demonstrate the comparison between experiment and theory over a more narrow separation interval from 100 to 150 nm where only each third experimental point is plotted together with its error bars $[\pm\Delta a, \pm\Delta^{\text{tot}}(\Delta F^{\text{expt}})]$ shown as crosses (the absorbed power is 8.5 mW). Note that these errors are entirely experimental. They are determined at a 95% confidence level as a combination of random and systematic errors and are not connected with any theory. The theoretical force differences ΔF^{theor} computed at $T = 300$ K with neglected and included dc conductivity in the dark phase are shown as the solid and dashed lines, respectively. From Fig. 2 one concludes that inclusion of the dc conductivity in the dark phase is excluded by the experimental data at a 95% confidence level within the separation region from 100 to 150 nm.

Note that computations of the Casimir force between an Au coated sphere and Si plate were done using the proximity force approximation [26]

$$F(a, T) = 2\pi R\mathcal{F}(a, T), \quad (15)$$

where $\mathcal{F}(a, T)$ is given by (1). The dielectric permittivities $\varepsilon^{(1)}(i\xi)$ and $\varepsilon^{(2)}(i\xi)$ for Si and Au, respectively, were obtained from the tabulated optical data [23] by means of the Kramers-Kronig relations. Practically the same results can be obtained using the analytical representations for $\varepsilon^{(n)}(i\xi)$. For Au high precision six-oscillator model is considered in [7]. For Si, the imaginary part of the dielectric permittivity can be modeled as a constant within some frequency region [27]

$$\text{Im } \varepsilon^{(1)}(\omega) = \begin{cases} \bar{\varepsilon}, & \omega_0 \leq \omega \leq \omega_1, \\ 0, & \omega < \omega_0, \omega > \omega_1, \end{cases} \quad (16)$$

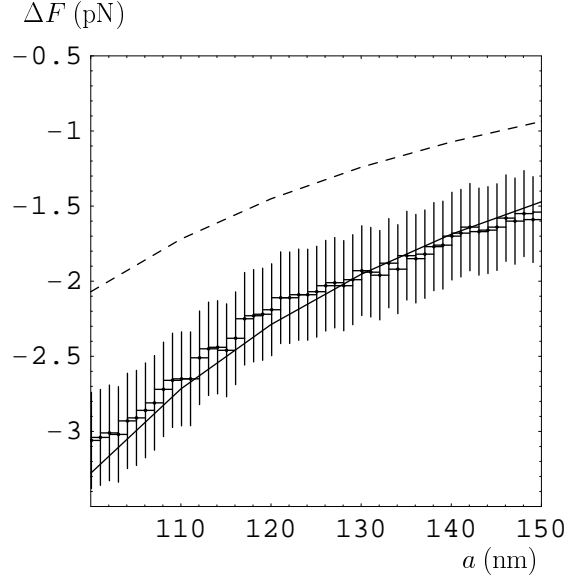


Figure 2. Mean experimental differences of the Casimir forces with their experimental errors are shown as crosses. Solid and dashed lines represent the theoretical differences computed at $T = 300\text{K}$ using the model of dielectric permittivity of high-resistivity Si neglecting and including the dc conductivity, respectively.

where $\omega_0 = 3.22\text{ eV}$, $\omega_1 = 4.62\text{ eV}$, $\bar{\epsilon} = 48$. Using the Kramers-Kronig relation, this results in

$$\epsilon^{(1)}(i\xi) = 1 + \frac{\bar{\epsilon}}{\pi} \ln \left(\frac{\omega_1^2 + \xi^2}{\omega_0^2 + \xi^2} \right). \quad (17)$$

The use of the approximation (17) instead of the tabulated optical data [23] leads to less than 1% deviations in the Casimir force (15) between an Au sphere and Si plate. At separations $a = 50, 100, 200, 400$ and 600 nm this deviation is equal to -0.58% , 0.30% , 0.70% , 0.84% and 0.86% , respectively.

Another analytic formula representing the dielectric permittivity of Si along the imaginary frequency axis takes the form [28]

$$\epsilon^{(1)}(i\xi) = \epsilon^{(1)}(i\infty) + \frac{[\epsilon^{(1)}(i0) - \epsilon^{(1)}(i\infty)]\omega_0^2}{\omega_0^2 + \xi^2}, \quad (18)$$

where $\omega_0 = 6.6 \times 10^{15}\text{ rad/s}$, $\epsilon^{(1)}(i0) = 11.87$ and $\epsilon^{(1)}(i\infty) = 1.035$. This representation also leads to less than 1% differences in the case of Au sphere above Si plate in comparison to computations using the tabulated optical data for Si. Thus, at separations $a = 50, 100, 200, 400$ and 600 nm the relative deviations between both computations are equal to 0.75% , 0.74% , 0.62% , 0.55% and 0.52% . Because of this, both the tabulated optical data for Si and different analytical representations for its dielectric permittivity can be used in the computations within 1% precision.

Another important experimental test is the measurement of the thermal Casimir-Polder force through center-of-mass oscillations of the trapped Bose-Einstein condensate [16]. In this experiment, not only were different models of the dielectric response compared with the data [16,29], but the thermal effect was measured for the first time in Casimir physics. The condensate of Rb atoms was at a distance from 7 to $11\text{ }\mu\text{m}$ from a fused SiO_2 dielectric substrate. In the

original publication this substrate was considered as a dielectric material with the static dielectric permittivity $\varepsilon^{(1)}(0) = 3.81$. However, at room temperature (in an equilibrium situation the temperature of the substrate and environment were equal to $T_S = T_E = 310$ K) SiO_2 has nonzero conductivity which is ionic in nature and varies from 10^{-9} s^{-1} to 10^2 s^{-1} depending on the concentration of alkali ions which are always present as trace constituents [30,31]. This conductivity was taken into account in [29] in accordance with (12).

The directly measured quantity in the experiment [16] was the relative shift of the frequency of center-of-mass oscillations of the Bose-Einstein condensate due to the presence of the Casimir-Polder force

$$\gamma_z = \frac{|\omega_0 - \omega_z|}{\omega_0} = \frac{1}{2m\omega_0^2} |\langle \partial_z F^A(z, T) \rangle|. \quad (19)$$

Here, $m = 1.443 \times 10^{-25} \text{ kg}$ is the mass of the Rb atom, ω_0 is the unperturbed trap frequency $\omega_0 = 2\pi \times 229 \text{ rad/s}$, $F^A(z, T)$ in the equilibrium situation is given by the negative derivative of $\mathcal{F}^A(z, T)$ defined in (4) with respect to z , and the averaging is done over the period of condensate oscillations and over the volume of the condensate cloud.

The experimental data for the relative frequency shift γ_z are presented as crosses in Fig. 3. The arms of the crosses show the total experimental errors determined in [16] at a 67% confidence level individually for each data point. The results of theoretical computations using (19) with neglected dc conductivity of SiO_2 substrate are shown by the solid line [16,29]. As is seen in the figure, theory neglecting the dc conductivity of fused silica is in a very good agreement with the data. In the same figure, the results of theoretical computations by (19) with dc conductivity of SiO_2 included are shown by the dashed line [29]. As is seen in the figure, the dashed line is in evident disagreement with the first two experimental points.

The experiment was repeated for two more times in situations out of thermal equilibrium ($T_S = 479 \text{ K}$, $T_E = 310 \text{ K}$ and $T_S = 605 \text{ K}$, $T_E = 310 \text{ K}$), i.e., with the temperature of the plate higher than the environment temperature. In this case the Casimir-Polder force $F^A(z, T)$ in (19) should be replaced with

$$F^A(z, T_S, T_E) = F^A(z, T_E) + F^{\text{neq}}(z, T_S) - F^{\text{neq}}(z, T_E), \quad (20)$$

where the explicit form of the function $F^{\text{neq}}(z, T)$ can be found in [16,29,32].

In Fig. 4 the experimental data for γ_z are shown for (a) $T_S = 479 \text{ K}$ and (b) $T_S = 605 \text{ K}$ as crosses with their arms indicating the experimental errors. As in Fig. 3, the theoretical results

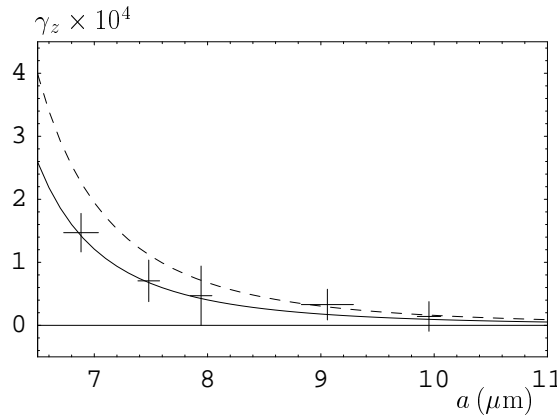


Figure 3. Fractional change in the trap frequency versus separation in thermal equilibrium with $T_S = T_E = 310 \text{ K}$ computed by neglecting (solid line) and including (dashed line) the conductivity of the dielectric substrate. The experimental data are shown as crosses.

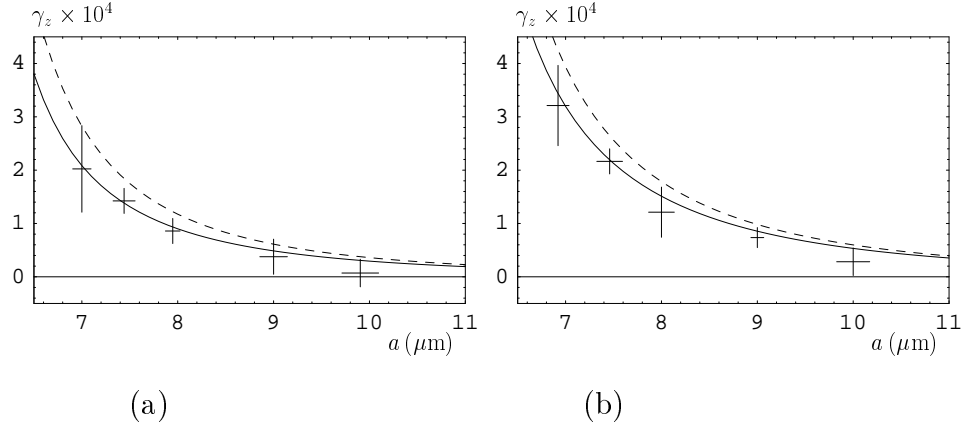


Figure 4. Fractional change in the trap frequency versus separation out of thermal equilibrium with (a) $T_S = 479$ K, $T_E = 310$ K and (b) $T_S = 605$ K, $T_E = 310$ K. Computations are done by neglecting (solid line) and including (dashed line) the conductivity of the dielectric substrate. The experimental data are shown as crosses.

computed with the dc conductivity of SiO_2 plate neglected and included are shown by the solid and dashed lines, respectively. As is seen in Fig. 4(a), the dashed line is in disagreement with the three experimental points, and the other two only touch it. In Fig. 4(b) all data points exclude the theoretical approach represented by the dashed line. Thus, the inclusion of the dc conductivity of the dielectric substrate into the Lifshitz theory leads to disagreement with the experimental data. The calculational results using the Lifshitz theory with the dc conductivity neglected are in very good agreement with the experimental data. We emphasize that the theoretical results obtained with dc conductivity included do not depend on the value of conductivity, but only of the fact that it is nonzero. Importantly, the inclusion of conductivity does not influence the nonequilibrium contributions to γ_z . Thus, the puzzling effect comes only from the contribution to γ_z given by the Lifshitz theory.

4. Phenomenological approach and its justification

In the above, both theoretical and experimental problems are elucidated arising in the Lifshitz theory when applied to dielectric materials at nonzero temperature with dc conductivity included. Recently, these problems were discussed also in [33] but no solution was found. In [33] only “weakly conducting” materials were considered which possess nonzero conductivity at zero temperature. These are in fact metallic-type semiconductors with a dopant concentration above critical. To satisfy the Nernst heat theorem, it was suggested that there is nonzero residual relaxation at $T = 0$, i.e., the crystal lattice is not perfect. Materials of such type have nothing to do with dielectrics whose conductivity vanishes when temperature goes to zero. The formalism of [33] is also in contradiction with precision experiments dealing with metallic test bodies [6,7].

To avoid contradictions of the Lifshitz theory with the principles of thermodynamics and experimental results, one can propose the following phenomenological prescription. In the applications of the Lifshitz theory to all materials having zero conductivity at zero temperature, the presence of free charge carriers at nonzero temperature should be disregarded. The dielectric permittivity of such materials should be described by equation (5). In the applications of the Lifshitz theory to all materials having nonzero conductivity at zero temperature, free charge carriers should be included in the framework of the generalized plasma-like model

$$\varepsilon_{\text{gp}}(i\xi) = 1 + \frac{\omega_p^2}{\xi^2} + \sum_j \frac{g_j}{\omega_j^2 + \xi^2 + \gamma_j \xi}, \quad (21)$$

where all $\omega_j \neq 0$. This prescription is consistent with thermodynamics and all available

experimental data [22,24,25,34].

The physical reasons of why the Lifshitz theory does not allow the inclusion of real conductivity processes can be understood as follows. Lifshitz derived his famous formulas under the condition of thermal equilibrium. This means that not only $T = \text{const}$, but also all irreversible processes connected with the dissipation of energy into heat have already been terminated [35,36]. The Drude-like dielectric function (12) is derived from the Maxwell equations with a real drift current of conduction electrons $\mathbf{j} = \sigma_0 \mathbf{E}$ initiated by the external electric field \mathbf{E} [24]. The drift current is an irreversible process which takes a system out of thermal equilibrium. This current leads to Joule heating of the Casimir plates (Ohmic losses) [37]. To preserve the temperature constant, one should admit that there exists an unidirectional flux of heat from the medium to the heat reservoir [38]. Such interactions between a system and a heat reservoir are prohibited by the definition of thermal equilibrium [39]. One can conclude that the substitution of the Drude-like dielectric permittivities into the Lifshitz formulas violate the applicability conditions under which this theory was developed. Then it is not surprising that such substitution results in problems with thermodynamics and contradictions with experimental data. From this point of view, the Lifshitz theory is free of problems and paradoxes which arise only if it is applied incorrectly.

Another question to be answered is the following. One could agree with the fact that the conductivity processes in real materials connected with the drift current of conduction electrons violate the applicability conditions of the Lifshitz theory and, thus, are not described by this theory. These processes, however, exist in reality. Because of this should they be taken into account in some future general theory of dispersion forces? At the moment we have no answer to this question. All computations on the basis of the Lifshitz theory for dielectrics with dc conductivity neglected and for metals by using the plasma-like dielectric permittivity (21) are in agreement with both thermodynamics and experimental data. This suggests that the conductivity properties are not related to dispersion forces. More precise experiments will probably bring additional information to bear on this subject.

5. Approaches based on the modification of reflection coefficients due to screening effects

Recently it was proposed [17,18] that the problems of the Lifshitz theory described above may have an alternative solution if the screening effects due to free charge carriers are taken into account. This approach includes both the drift and diffusion currents of free charge carriers through use of the transport Boltzmann equation [18]. It uses the standard Lifshitz formulas (1) and (4) for the free energy of wall-wall and atom-wall interaction with the TE reflection coefficient, \tilde{r}_{TE} , as defined in (2) within the Drude model approach, but with the modified TM reflection coefficient

$$\tilde{r}_{\text{TM}}(i\xi, k_{\perp}) = \frac{\tilde{\varepsilon}(i\xi)q - k - \frac{k_{\perp}^2}{\eta(i\xi)} \frac{\tilde{\varepsilon}(i\xi) - \varepsilon(i\xi)}{\varepsilon(i\xi)}}{\tilde{\varepsilon}(i\xi)q + k + \frac{k_{\perp}^2}{\eta(i\xi)} \frac{\tilde{\varepsilon}(i\xi) - \varepsilon(i\xi)}{\varepsilon(i\xi)}}. \quad (22)$$

Here,

$$\tilde{\varepsilon}(i\xi) = \varepsilon(i\xi) + \frac{\omega_p^2}{\xi(\xi + \gamma)}, \quad k^2 = k_{\perp}^2 + \tilde{\varepsilon}(i\xi) \frac{\xi^2}{c^2}, \quad (23)$$

where $\varepsilon(i\xi)$ is the permittivity of the bound core electrons given in (5). The quantity $\eta(i\xi)$ is defined as

$$\eta(i\xi) = \left[k_{\perp}^2 + \kappa^2 \frac{\varepsilon(0)}{\varepsilon(i\xi)} \frac{\tilde{\varepsilon}(i\xi)}{\tilde{\varepsilon}(i\xi) - \varepsilon(i\xi)} \right]^{1/2}, \quad (24)$$

where $1/\kappa$ is the screening length. If the charge carriers of density n are described by the classical Maxwell-Boltzmann statistics, as assumed in [18], one gets the Debye-Hückel screening length

[40,41]

$$R_D = \frac{1}{\kappa} = \left(\frac{\varepsilon_0 k_B T}{4\pi e^2 n} \right)^{1/2}. \quad (25)$$

Assuming Fermi-Dirac statistics, one arrives at the Thomas-Fermi screening length [40,41]

$$R_{TF} = \frac{1}{\kappa} = \left(\frac{\varepsilon_0 E_F}{6\pi e^2 n} \right)^{1/2}. \quad (26)$$

Here, e is the electron charge, $\varepsilon_0 = \varepsilon(0)$ is the dielectric constant due to the core electrons, and $E_F = \hbar\omega_p$ is the Fermi energy.

Note that the paper [18] applies this approach to intrinsic semiconductor media. Bearing in mind, however, that the classical Drude dielectric permittivity is exploited, it is justified to apply it for metals and doped semiconductors (both dielectric and metallic type) as well, by substituting the respective screening length defined by (25) or (26).

Now we consider whether or not the approach taking the screening effects into account is consistent with thermodynamics. We concentrate our attention on the case of dielectric materials whose conductivity goes to zero when the temperature vanishes. We start from the case of two similar dielectric semispaces $[\varepsilon^{(1)} = \varepsilon^{(2)} = \bar{\varepsilon}]$. The Casimir free energy per unit area is given by (1) with the reflection coefficients $r_{TE}^{(1)} = r_{TE}^{(2)} = \tilde{r}_{TE}$ from (2), (23), and $r_{TM}^{(1)} = r_{TM}^{(2)} = \tilde{r}_{TM}$ from (22).

The derivation of the asymptotic behavior of the Casimir free energy and entropy at low temperature is performed in perfect analogy with the derivation of (7) and (13) in [5]. First we introduce the small parameter ($\xi \geq \xi_1$)

$$\beta = \frac{4\pi\sigma(T)}{\xi}, \quad (27)$$

which goes to zero due to (11) when the temperature vanishes. Then we expand the reflection coefficients $\tilde{r}_{TM,TE}$ at all nonzero Matsubara frequencies in powers of the parameter β

$$\begin{aligned} \tilde{r}_{TM}(i\zeta, y) &= r_{TM}(i\zeta, y) + \beta \frac{\varepsilon y[2y^2 + (\varepsilon - 2)\zeta^2]}{\sqrt{y^2 + (\varepsilon - 1)\zeta^2}[\varepsilon y + \sqrt{y^2 + (\varepsilon - 1)\zeta^2}]^2} + O(\beta^2), \\ \tilde{r}_{TE}(i\zeta, y) &= r_{TE}(i\zeta, y) + \beta \frac{y[y - \sqrt{y^2 + (\varepsilon - 1)\zeta^2}]}{\sqrt{y^2 + (\varepsilon - 1)\zeta^2}[y + \sqrt{y^2 + (\varepsilon - 1)\zeta^2}]} + O(\beta^2). \end{aligned} \quad (28)$$

Here, the reflection coefficients $r_{TM,TE}$ are defined by (2) with the dielectric permittivity (5). They lead to the asymptotic behavior of the Casimir free energy per unit area given in (7). The dimensionless variables ζ and y are connected with other dimensional ones by the equations

$$y = 2aq, \quad \zeta = \frac{\xi}{\omega_c} = \frac{2a\xi}{c}, \quad (29)$$

where $q = q(\xi)$ is defined in (3).

Now we substitute (28) in (1) and arrive at the following expression for the Casimir free energy per unit area taking the screening effects into account

$$\tilde{\mathcal{F}}(a, T) = \mathcal{F}(a, T) + \frac{k_B T}{16\pi a^2} \left\{ \int_0^\infty y dy \ln \left[1 - \tilde{r}_0^2(y) e^{-y} \right] + \text{Li}_3(r_0^2) + Q(T) \right\}, \quad (30)$$

where

$$\tilde{r}_0(y) \equiv \frac{\varepsilon_0 \sqrt{y^2 + (2a\kappa)^2} - y}{\varepsilon_0 \sqrt{y^2 + (2a\kappa)^2} + y} \quad (31)$$

and $Q(T)$ vanishes exponentially when $T \rightarrow 0$. The free energy $\mathcal{F}(a, T)$ in (30) is given by (7), and κ in (31) is defined in (25). Calculating the negative derivative of both sides of (30) with respect to T , we obtain the asymptotic behavior of the Casimir entropy at low temperature

$$\begin{aligned} \tilde{S}(a, T) = S(a, T) - \frac{k_B}{16\pi a^2} & \left\{ \int_0^\infty y dy \ln [1 - \tilde{r}_0^2(y)e^{-y}] + \text{Li}_3(r_0^2) \right. \\ & \left. - 8a^2 \varepsilon_0 T \frac{\partial \kappa^2}{\partial T} \int_0^\infty dy \frac{y^2 \tilde{r}_0(y)}{e^y - \tilde{r}_0^2(y)} \frac{1}{\sqrt{y^2 + (2a\kappa)^2} [\varepsilon_0 \sqrt{y^2 + (2a\kappa)^2} + y]^2} + Q(T) + TQ'(T) \right\}, \end{aligned} \quad (32)$$

where $S(a, T)$ is defined in (10). It can be easily seen that the last three terms in the curly brackets on the right-hand side of this equation go to zero when T goes to zero for any dielectric material.

The behavior of the first two terms in the curly brackets on the right-hand side of (32) when T goes to zero is more involved. If $n(T)$ exponentially decays to zero with vanishing temperature (as is true for pure insulators and intrinsic semiconductors), then according to (25) so does κ . As a result, $\tilde{r}_0(y) \rightarrow r_0 = (\varepsilon_0 - 1)/(\varepsilon_0 + 1)$ and the first two terms in the curly brackets cancel. Then the Casimir entropy $\tilde{S}(a, T)$ goes to zero when T vanishes following the same law as $S(a, T)$, i.e., as T^2 . This means that for insulators and intrinsic semiconductors the formalism under consideration is in agreement with the Nernst heat theorem.

However, there is a wide class of dielectric materials (such as doped semiconductors with dopant concentration below critical and solids with ionic conductivity) for which n does not go to zero when T goes to zero. In fact, conductivity can be presented as [41]

$$\sigma(T) = n |e| \mu, \quad (33)$$

where μ is the mobility of charge carriers. Although $\sigma(T)$ goes to zero exponentially fast for all dielectrics when T goes to zero, for most of them this happens due to the vanishing mobility. For instance, as mentioned in Sec. 3, the conductivity of SiO_2 used as the plate material in the experiment [16] is ionic in nature and is determined by the concentration of impurities. For all such materials, in accordance with (25), $\kappa \rightarrow \infty$ when $T \rightarrow 0$. As a result, $\tilde{r}_0(y) \rightarrow 1$ when T goes to zero in accordance with (31). In this case we obtain from (32)

$$\tilde{S}(a, 0) = \frac{k_B}{16\pi a^2} [\zeta(3) - \text{Li}_3(r_0^2)] > 0 \quad (34)$$

in violation of the Nernst heat theorem. This means that for a wide class of dielectric materials the proposed approach taking the screening effects into account is thermodynamically inconsistent in the same way as the standard Lifshitz theory with the dc conductivity included [compare with (14)].

We emphasize that the existence of dielectric materials for which n does not go to zero but μ does go to zero when T vanishes demonstrates that the reflection coefficient (22) at $\xi = 0$ is ambiguous. In reality, for such materials $\tilde{r}_{\text{TM}}(0, k_\perp) \rightarrow 1$ when T and μ simultaneously vanish. This is because $\kappa \rightarrow \infty$ when $T \rightarrow 0$ in disagreement with physical intuition that there should be no screening at zero mobility. This ambiguity is connected with the break of continuity of the reflection coefficient $\tilde{r}_{\text{TM}}(i\xi, k_\perp)$ at the point $\xi = 0, T = 0$. If one takes the limit $T \rightarrow 0$ first, keeping $\xi = \text{const} \neq 0$, the standard Fresnel reflection coefficients r_{TM} from (2) with no screening is reproduced. This property is preserved in the subsequent limiting transition $\xi \rightarrow 0$. As was noted in [42] on general grounds, the violation of the Nernst heat theorem is caused by the breaks in the continuity of the reflection coefficients at the origin of the (ξ, T) -plane.

The theoretical approach taking the screening effects into account can also be applied to describe the Casimir-Polder atom-wall interaction. In this case it is also inconsistent with thermodynamics for dielectric materials with a nonvanishing density of charge carriers. To find the asymptotic behavior of the free energy at low temperature, one should substitute into (4) the reflection coefficients $r_{\text{TM,TE}}^{(1)} = \tilde{r}_{\text{TM,TE}}$, as defined in (28). The result is

$$\tilde{\mathcal{F}}^A(a, T) = \mathcal{F}^A(a, T) - \frac{k_B T \alpha(0)}{8a^3} \left[\int_0^\infty y^2 dy \tilde{r}_0(y) e^{-y} - 2r_0 + Q_1(T) \right], \quad (35)$$

where the asymptotic behavior of $\mathcal{F}^A(a, T)$ is presented in (8), and $Q_1(T)$ vanishes exponentially when $T \rightarrow 0$. Calculating the negative derivative of both sides of (35) with respect to T , one obtains

$$\begin{aligned} \tilde{S}^A(a, T) = S^A(a, T) + \frac{k_B \alpha(0)}{8a^3} & \left[\int_0^\infty y^2 dy \tilde{r}_0(y) e^{-y} - 2r_0 \right. \\ & \left. + 4a^2 \varepsilon_0 T \frac{\partial \kappa^2}{\partial T} \int_0^\infty dy \frac{y^3 e^{-y}}{\sqrt{y^2 + (2a\kappa)^2} (\varepsilon_0 \sqrt{y^2 + (2a\kappa)^2} + y)^2} + Q_1(T) + T Q_1'(T) \right], \end{aligned} \quad (36)$$

where $S^A(a, T)$ is defined in (10). For dielectric materials with exponentially decaying $n(T)$ in the limit of low T , $\tilde{r}_0(y) \rightarrow r_0$ and all terms on the right-hand side of (36) added to $S^A(a, T)$ go to zero when $T \rightarrow 0$. As a result, $\tilde{S}^A(a, T)$ goes to zero as T^3 and the Nernst theorem is satisfied. On the contrary, for all dielectric materials with nonvanishing n we get $\tilde{r}_0(y) \rightarrow 1$ when $T \rightarrow 0$ and in this case (36) results in

$$\tilde{S}^A(a, 0) = \frac{k_B \alpha(0)}{4a^3} (1 - r_0) > 0. \quad (37)$$

This means that the Nernst theorem is violated in the same way as in (14) for the standard Lifshitz theory with the dc conductivity included.

The theoretical predictions following from the approach proposed in [18] can be compared with the measurement data of the experiment on the modulation of the Casimir force with light (see Sec. 3 for details). In Fig. 5(a) and 5(b) we present the experimental data of this experiment obtained for the absorbed powers P_{w1} and P_{w2} , respectively, with the experimental errors of force measurements determined at 70% confidence level. The theoretical difference Casimir force, computed using the standard Lifshitz theory with the dc conductivity of high-resistivity Si in the dark phase neglected, is shown by the solid lines. The theoretical difference Casimir force computed by including the screening effects at all Matsubara frequencies, as described above, is shown by the dashed lines. It can be seen that the standard Lifshitz theory is consistent with data, whereas the theoretical approach taking the screening effects into account is excluded by the data at a 70% confidence level.

In Sec. 4 the contradictions with thermodynamics and the experimental data of the standard Lifshitz theory with the dc conductivity included are explained based on the violation of its applicability condition, i.e., of thermal equilibrium, in the presence of drift current. In a similar way, the approach taking the screening effects into account applies the Lifshitz theory to physical phenomena involving both the drift and diffusion currents. The latter is caused by the nonequilibrium distribution of charge carriers in an external field, i.e., by a physical situation out of thermal equilibrium. This is because the diffusion current is determined by a nonzero gradient of charge carrier density, whereas for homogeneous systems in thermal equilibrium the charge carrier density must be homogeneous. Thus, the problems and paradoxes arising in this case are also explained by the use of the Lifshitz theory outside of its defined application region.

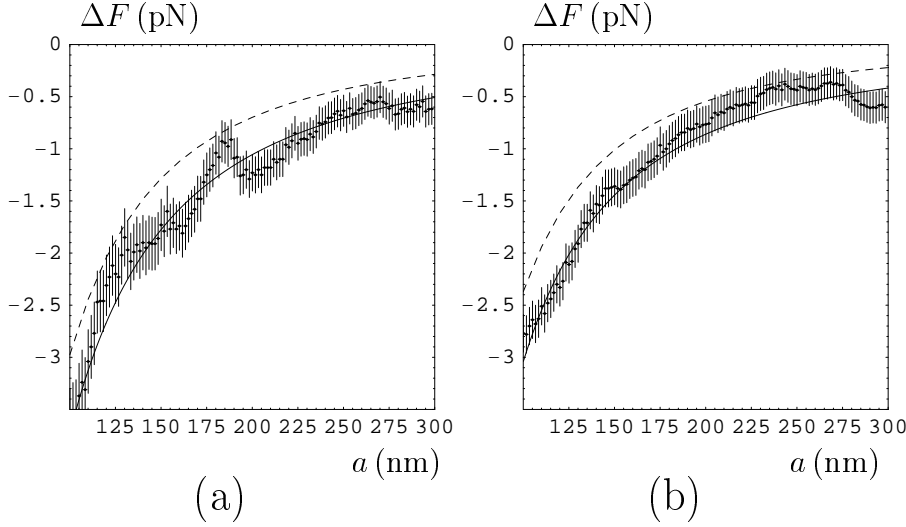


Figure 5. Differences of the Casimir forces between an Au-coated sphere and a Si plate, in the presence and in the absence of laser light on the plate, versus separation for the absorbed power of (a) 9.3 mW and (b) 4.7 mW. The experimental data are shown as crosses. Solid and dashed lines indicate the theoretical results calculated using the standard Lifshitz theory with the dc conductivity of Si in the dark phase neglected, and the generalization of this theory [17,18], respectively.

An earlier attempt to incorporate the screening effects in the theory of dispersion forces was undertaken in [17]. In this approach the effect of screening was taken into account only for the static field in the interaction of an atom with a dielectric wall. Bearing in mind that in accordance with (4) the TE reflection coefficient at zero frequency does not contribute to the result, the single modified coefficient was [17]

$$r_{\text{TM}}^{\text{mod}}(0, k_{\perp}) = \frac{\varepsilon_0 \sqrt{k_{\perp}^2 + \kappa^2} - k_{\perp}}{\varepsilon_0 \sqrt{k_{\perp}^2 + \kappa^2} + k_{\perp}}, \quad (38)$$

where κ is given in (25). It is easily seen that (38) coincides with the general TM reflection coefficient $\tilde{r}_{\text{TM}}(i\xi, k_{\perp})$ defined in (22) if one puts $\xi = 0$.

The reflection coefficient (38) can be obtained by the formal replacement of the dielectric permittivities depending only on the frequency with the permittivities depending both on frequency and the wave vector in the Fresnel reflection coefficient of an uniaxial crystal. To confirm that this is the case, one should introduce nonisotropic permittivities $\varepsilon_x = \varepsilon_y \neq \varepsilon_z$ and replace the coefficients r_{TM} and r_{TE} in (2) with

$$r_{\text{TM}}^{\text{mod}}(i\xi, k_{\perp}) = \frac{\sqrt{\varepsilon_x(i\xi)\varepsilon_z(i\xi)}q - k_z}{\sqrt{\varepsilon_x(i\xi)\varepsilon_z(i\xi)}q + k_z}, \quad r_{\text{TE}}^{\text{mod}}(i\xi, k_{\perp}) = \frac{q - k_x}{q + k_x}. \quad (39)$$

Here, k_z and k_x are defined by (3) with the replacement of $\varepsilon(i\xi)$ by $\varepsilon_z(i\xi)$ and $\varepsilon_x(i\xi)$, respectively. For ε_x and ε_z depending only on the frequency, (39) is commonly used for the description of uniaxial crystals (see, e.g., [43,44]). Then (38) follows from (39) if one puts

$$\varepsilon_x(0) = \varepsilon_0, \quad \varepsilon_z(0) = \varepsilon_z(0, k_{\perp}) = \varepsilon_0 \left(1 + \frac{\kappa^2}{k_{\perp}^2} \right), \quad (40)$$

where now $\varepsilon_z(0)$ depends on the wave vector, i.e., spatial dispersion is present. Thus, to obtain (38) one substitutes into the Fresnel reflection coefficient (39) derived from the standard continuity boundary conditions a dielectric permittivity depending on the wave vector (see [45,46] why this is inappropriate).

We emphasize that the general reflection coefficients taking the screening effects into account [the transverse electric from (2) with the dielectric permittivity (23) and the transverse magnetic from (22)] cannot be represented in the form of (39). To prove this statement, we note that from the equality $\tilde{r}_{\text{TE}} = r_{\text{TE}}^{\text{mod}}$ it follows that $\tilde{\varepsilon}(i\xi) = \varepsilon_x(i\xi)$. Thus, in accordance with (23), $\varepsilon_x(i\xi) \rightarrow \infty$ when $\xi \rightarrow 0$. Keeping in mind that $\varepsilon_z(0) > 0$, from (39) one obtains $r_{\text{TM}}^{\text{mod}}(0, k_{\perp}) = 1$. This is in contradiction with the above result that at zero frequency the reflection coefficient (22) coincides with the reflection coefficient (38). This means that the general approach taking the screening effects into account [18] cannot be described in terms of any dielectric permittivity which includes spatial dispersion.

The approach based on the modification of only one reflection coefficient (38) meets the same thermodynamical difficulties as the general approach considered above. Thus, if one computes the Casimir entropy with the reflection coefficient (38) and all the other standard ones, as given by (2), it goes to zero when the temperature vanishes for dielectrics with exponentially vanishing charge carrier density n . However, for dielectrics with nonvanishing n the result (37) is reproduced, i.e., the Nernst heat theorem is violated [20].

The modification of the TM reflection coefficient in accordance with (38) can be verified experimentally by using the measurement data of the optical modulation experiment discussed in Sec. 3. In theoretical computations at zero frequency the expression (38) was used. At all nonzero Matsubara frequencies the standard reflection coefficients (2) were substituted into the Lifshitz formula. The computational results using the standard reflection coefficients at all nonzero Matsubara frequencies coincide with those obtained with the general reflection coefficients $\tilde{r}_{\text{TM}}(i\xi, k_{\perp})$ and $\tilde{r}_{\text{TE}}(i\xi, k_{\perp})$. Then we return to the dashed lines in Fig. 5(a,b) representing the difference Casimir force computed with the inclusion of screening effects. More generally, it can be easily seen that for materials with sufficiently small charge carrier density (for instance for Si with $n \leq 10^{17} \text{ cm}^{-3}$) the general reflection coefficient (22) including the screening effects leads to approximately the same values at room temperature, as the standard one at all nonzero Matsubara frequencies. Thus, the theoretical results computed by using the modified TM reflection coefficient (38) are excluded by data at a 70% confidence level.

6. Conclusions and discussion

In the above we have considered the application of the Lifshitz theory to real metals, dielectrics and semiconductors. When the conductivity processes, connected with the presence of free charge carriers, are included into the model of the dielectric response, problems and paradoxes in the application of the Lifshitz theory are immediately apparent. They are manifested in the contradictions of the Lifshitz theory with both thermodynamics and experiment. We have listed several measurements performed by different experimental groups with metals, dielectrics and semiconductors which are in contradiction with the theoretical results taking into account the drift current of conduction electrons.

In searching for reasons of why the problems and paradoxes arise, it was emphasized that the Lifshitz theory was formulated for systems in thermal equilibrium. As a result, this theory is not applicable to physical systems where processes go on which violate thermal equilibrium. Specifically, in thermal equilibrium all processes of heat transfer, diffusion and others connected with the dissipation of energy are terminated. A question then arises if the drift current of charge carriers which leads to a unidirectional flux of Joule heat from the Casimir plates to the heat reservoir is compatible with the state of thermal equilibrium. Our conclusion is that it is not compatible with thermal equilibrium and, thus, cannot be considered in the framework of

the Lifshitz theory. The inclusion of conductivity properties in the Lifshitz theory is in violation of its basic applicability condition.

We have analyzed recent attempts to solve problems and paradoxes using screening effects and diffusion processes. We have shown that these attempts are in conflict with thermodynamics and in contradiction with experiment. This is because diffusion processes make the deviations from thermal equilibrium even more dramatic.

One may conclude that the Lifshitz theory, if used within its application region, is free of problems and paradoxes. Problems and paradoxes arise when the basic applicability condition of the Lifshitz theory, thermal equilibrium, is not taken into account by using the Drude model or introducing the diffusion current. What is more, phenomenologically the Lifshitz theory leads to both thermodynamically and experimentally consistent results if the processes of electric conductivity connected with both drift and diffusion currents are simply disregarded. Future progress should show if these processes have any relation to the phenomenon of dispersion forces.

Acknowledgments

This work was supported by Deutsche Forschungsgemeinschaft, Grant No 436 RUS 113/789/0–4. The author is grateful to the Center of Theoretical Studies and Institute of Theoretical Physics of Leipzig University, where this work was performed, for their kind hospitality.

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